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THEORETICAL CONSIDERATIONS ON THE EFFECTIVENESS OF CARBON SEEDING

by

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March 1962

U. S. ARMY SIGNAL RESEARCH AND DEVELOPMENT LABORATORY FORT MONMOUTH, NEW JERSEY

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ABSTRACT

Presented in the report are: some results of machine computations of the scattering functions, the extinction-, scattering-, and absorption-coefficients for pure absorbing carbon particles, and the compound carbon particles with concentric water shell of varying thickness; and an analysis of the absorption properties of various single particles.

The scattering and absorption properties of single particles are used to calculate the absorptivity, transmissivity, and reflectivity of three cloud models: a pure carbon particle cloud, a cloud of water droplets and carbon particles, and a cloud of compound concentric carbonwater particles. All three clouds are 1,000 meters thick and near the ground.

The temperature change of one particle caused by radiation absorption has been computed, and the total energy for evaporation of a cloud mass has been estimated.

From these calculations it can be concluded that a total carbon mass of the order of several hundred pounds per km will be necessary to evaporate a stratus-type cloud.

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THEORETICAL CONSIDERATIONS ON THE EFFECTIVENESS OF CARBON SEEDING

INTRODUCTION

The first big step in artificial cloud modification was taken in 1946 when, through experiments conducted by Schaeferl and Vonnegut, the artificial stimulation of precipitation from supercooled clouds became feasible. The technique applied by the two experimentalists was based on Findeisen's theory that in supercooled clouds the introduction of nuclei could initiate the formation of precipitation particles. Schaeferl found that small pieces of dry ice could form a large number of small ice crystals in such a cloud, and Vonnegut discovered that, also, tiny crystals of silver iodide would be very active as sublimation nuclei. Many successful seeding experiments with supercooled cumulus clouds as well as supercooled stratus clouds have since been carried out.

However, both of the above-mentioned seeding methods are confined to the application for supercooled clouds only, whose temperature is below 0°C; and all attempts to find a reliable method for the dissipation of warm clouds and fogs have been unsuccessful. The principal difficulty with warm-cloud seeding is that there is no "latent energy" in the cloud available which, after liberation, could carry on the dissipation process by itself.

In the case of supercooled clouds, this latent energy is the energy of fusion which becomes free after seeding and which has been provided in an indirect way by the decrease of the entropy of the system by nature itself. Whatever energy for a precipitation process or evaporation process is needed in a warm cloud, therefore, has to be fully supplied from an outside source. As such an energy source, the radiation energy from the sun is, of course, the most apparent. There are other forms of energy which could possibly be used; for instance, electrostatic forces for dissipation of fogs and clouds. These, however, will not be discussed in this report.

Using the properties of carbon-black particles suspended in the atmosphere, several attempts have been made to absorb and emit sunlight, thus influencing, artificially, the energy processes in the atmosphere and especially in clouds. In recent years, several investigators have made estimates of the energy absorbed in carbon spheres and clouds of carbon particles; and various hypotheses have been developed on the processes which might occur in carbon-seeded clouds. However, all these computations of the energy absorbed by carbon particles have been based on the so-called bulk method which uses the equivalent solid-mass layer for its absorption calculations, or they were based on very rough estimates of the absorption cross section. Such results, of course, may be considerably different from exact solutions of the formulas of the Mie theory. 4

Actual carbon-seeding experiments have been carried out by Van Straten, Ruskin, Dinger, and Mastenbrook⁵ and by personnel of the Air Force Geophysics Research Directorate, but none of these experimental tests of carbon seeding gave a decisive answer to the question of the effectiveness of this method. It therefore seemed desirable to conduct some theoretical studies on the feasibility of carbon seeding, including an evaluation of the absorption properties of carbon-cloud elements according to the Mie theory and to find from this a sound basis for future experiments.

DISCUSSION

Absorption and Scattering Properties of Single Carbon and Carbon-Water Particles

As mentioned previously, for a comprehensive solution of the problem it is necessary to investigate the optical properties of pure carbon particles as well as compound carbon and water particles. The compound carbon-water particle, a carbon sphere with a concentric water shell around it, will be an idealization to some extent and will probably occur, in this form, only in the case of condensation of water around the carbon center. However, it is the only combination carbon-water droplet which can be treated theoretically. The exact formulas for such compound particles have been worked out by Aden and Kerker and are an extension of Mie's theory. They consider three different media separated by two boundary layers. With this theory, the absorption and scattering properties of pure spherical homogeneous particles with complex refractive index, and also of spherical particles with concentric shell of varying thickness can be computed. The following notations will be used:

 θ = Scattering angle (θ = 0 is forward scattering) a = Radius of the inner sphere (carbon particle)

b = Outer radius of the shell

= Wavelength of the incident light

m = n (l - n'i) = complex refractive index

 $\alpha = 2 \pi a / \lambda = \text{size parameter for a}$ = $2 \pi b / \lambda = \text{size parameter for b}$

b/a = Specific ratio.

According to Mie's theory and from Aden and Kerker's treatment, the following quantities are derived.

The cross sections for extinction and scattering,

$$Q_e = \frac{2}{\sqrt[p]{2}} \sum_{n=1}^{\infty} (2n + 1) \operatorname{Re}(a_n + b_n)$$
 (1)

$$Q_{s} = \frac{2}{\gamma^{2}} \sum_{n=1}^{\infty} (2n + 1) \cdot (|a_{n}|^{2} + |b_{n}|^{2}).$$
 (2)

The cross section for absorption,

$$Q_{\mathbf{g}} = Q_{\mathbf{e}} - Q_{\mathbf{g}}. \tag{3}$$

The absolute intensities,

$$i_1 = |I_1|^2$$
 and $i_2 = |I_2|^2$ (4)

with $I_1 = \sum_{n=1}^{\infty} \frac{(2n+1)}{n(n+1)} \left\{ a_n \pi_n + b_n \overline{c}_n \right\}$ (5)

$$I_{2} = \sum_{1}^{\infty} \frac{(2n+1)}{n(n+1)} \left\{ a_{n} \overline{l}_{n} + b_{n} \pi_{n} \right\}.$$
 (6)

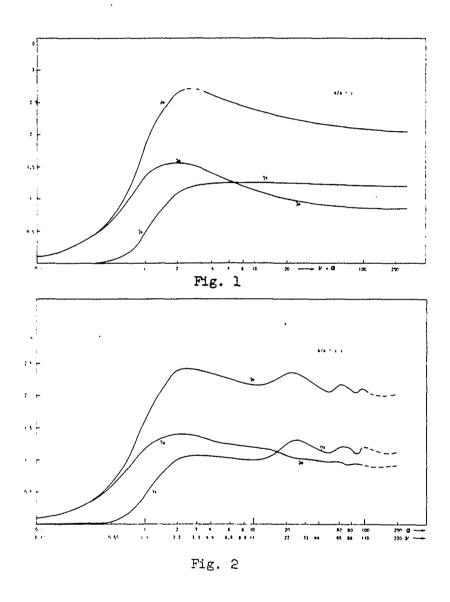
The functions a_n and b_n depend on the refractive index and the size parameter(s); the π_n and τ_n are functions of the scattering angle only.

Numerical computations have been carried out for Q_e , Q_s , i_1 , and i_2 for pure carbon particles, refractive index m=1.59-0.66i (b/a = 1), and carbon spheres with a concentric water shell (b/a variable) for size parameter values V from 0.1 to 200. The computations have been carried out under Signal Corps contract on the IBM 704 computer of the National Bureau of Standards, Washington, D. C.

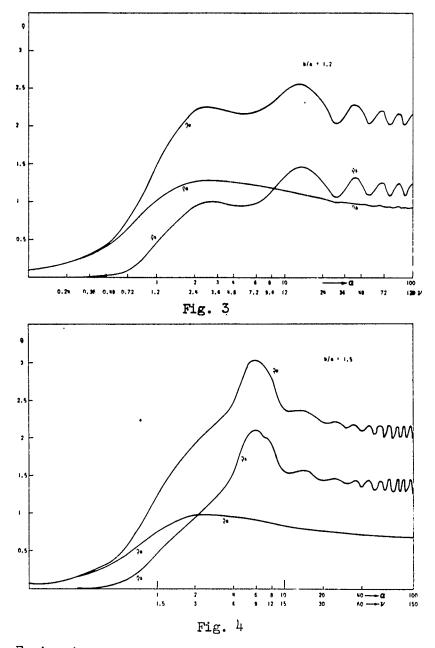
In Figs. 1 through 7 the computed total extinction-, scattering-, and absorption-coefficients for compound water-carbon particles for various ratios b/a from 1 to 0 are plotted as functions of the size parameter. Since, per definition, the Q_e , Q_g , and Q_a give the ratio between an area with which a particle interferes with the incident light and the actual geometrical cross section, it can be seen that the effective absorption cross section of a pure carbon particle (Fig. 1) for $1 \le \alpha \le 4$ becomes almost 1 1/2 its geometrical cross section. For $\alpha \le 0.5$ the extinction is practically due to absorptic only, and for $\alpha > 2$ scattering and absorption coefficients are of the same order of magnitude.

For absorbing particles, the Q-functions are very smooth and do not show the oscillations which are so characteristic for the extinction coefficient of nonabsorbing particles. However, as the thickness of the water shell around the carbon particle increases, more and more of the effect of the damping of the imaginary part of the refractive index is lost, and the oscillations in the Q-functions become stronger (b/a = 1.1, 1.2, 1.5, 2, 5 and 10 in Figs. 2 through 7, respectively) until for very large b/a ratios the Q-functions approach the values for pure water droplets.*

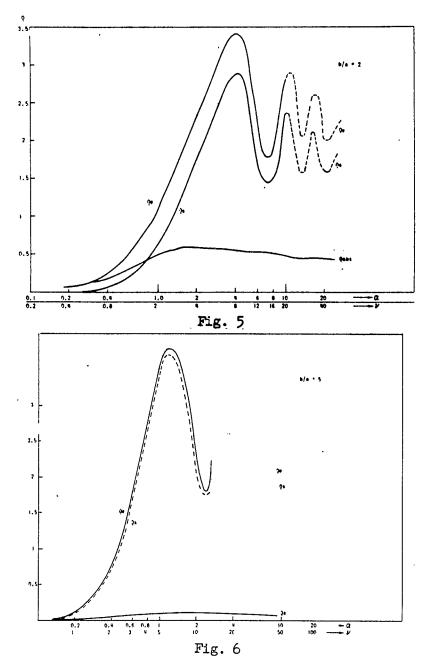
^{*}The curves in Figs. 5 through 7 for values $\mathcal V$ > 10 do not contain enough computed points to identify them unambiguously. Q_s ($\mathcal V$) for b/a - 10 is practically already equal to Q_s (\propto) for pure water.



Extinction-, scattering-, and absorption- coefficients for carbon particles of varying water shell thickness.



Extinction-, scattering-, and absorpt on-coefficients for carbon particles of varying water shell thickness.



Extinction-, scattering-, and absorption-coefficients for carbon particles of varying water shell thickness.

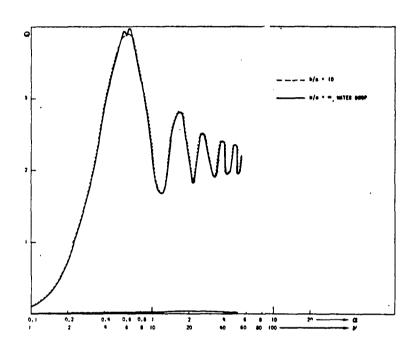


Fig. 7. Extinction-, scattering, and absorption-coefficients for carbon particles of varying water shell thickness.

In Fig. 8, the absorption coefficient Q_a (referring to a pure carbon particle as 100 percent), has been plotted as a function of b/a for constant total particle size ($\mathscr{V} = \text{const}$). It can be seen from this graph that for larger particles ($\mathscr{V} > 10$) the absorption coefficient of a carbon particle will be increased even by a thin water shell (b/a = 1.1). However, if b/a increases further, the water shell begins to reduce the absorption effect of the carbon nucleus. This shielding effect of the water shell is the stronger, the smaller the total particle size parameter.

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According to the definition for $\mathbb{Q}_{\mathbf{a}}$, the absorption cross section of one particle is

$$c_a = r^2 \cdot \mathcal{T} \cdot Q_a, \tag{7}$$

where r is the radius of the outer shell; i.e., in the case of compound particles, b is used instead of r. From this it follows that the absorption effect Ca of a compound particle which grows bigger and bigger disappears only if $Q_a(b)$ decreases more than b^2 increases with increasing b. However, since $\mathbb{Q}_{\mathbf{a}}$ is a function of $ext{$<$}$ and \mathscr{S} , or of either $ext{$<$}$ or \mathscr{S} plus b/a, different results are obtained for equation (7), depending on whether b/a or p^2 is kept constant. If α and θ grow proportionally (b/a = const), the increase of b2 will always be of higher order than the one for Qa, and therefore Ca \rightarrow oo as \mathscr{V} increases (Fig. 9). Whereas, if α is kept constant and the water is allowed to grow around the carbon particle (i.e., b/a variable), Q (\mathcal{P}) will decrease much more for increasing \mathcal{P} (Figs. 10 and 11). In Fig. 8 it is seen that even the efficiency factor for absorption Q_a itself increases above the one for a pure carbon particle if a thin water shell forms around the carbon core. In Figs. 10 and 11 for α = 0.1 and α = 1, the function $C_a = b^2 \mathcal{T}$ $C_a(b/a)$ has been plotted. A normalizing factor C has been added, so that C \cdot $b^2 \cdot \mathcal{T}$ $C_a(b/a = 1) = C_a(b/a = 1)$, or since in this case $b^2 = a^2$, C becomes $1/a^2$, which is constant per definition. C_a increases to a peak value two to three times the Ca for a pure carbon particle, and then diminishes again. This means that the water skin acts in the direction to increase the absorption effect of the carbon particle. The absolute values decrease considerably if the size of the carbon particle itself gets smaller. Computations up to specific ratios b/a of 150 show that the absorption cross section $C_a = b^2 \mathcal{T} \cdot Q_a$ remains finite and its obsolete value stays constant around the value for $C_a(B/a = 20)$.

An investigation was made of the wavelength dependence of the absorption coefficient of carbon particles. From Fig. 1 it can be seen that for \propto < 1.5 and for 3< < < 20 the absorption for carbon particles of constant size is wavelength dependent, and that for \propto < 1.5 the absorption is the smaller, the larger the wavelength. The opposite is the case for 3< \propto < 20. The range < < 1.5 corresponds for visible light to very small particles (r < 0.1,%). Over the wavelength range 0.3 < < < < < < < < 0.7, Qa can vary by a factor 2. This results in the phenomena that industrial smoke quite often appears red-brown in the transmitted light if the particles are uniform enough in size and very small. As the particles become less absorbing (Figs. 2 through 7), the slope of Qa becomes flatter and the wavelength dependence gradually disappears.

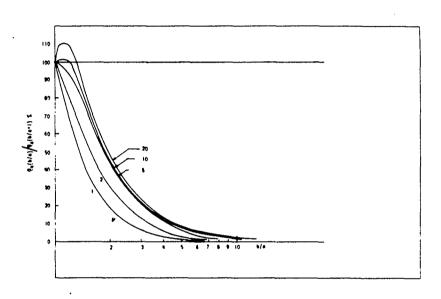
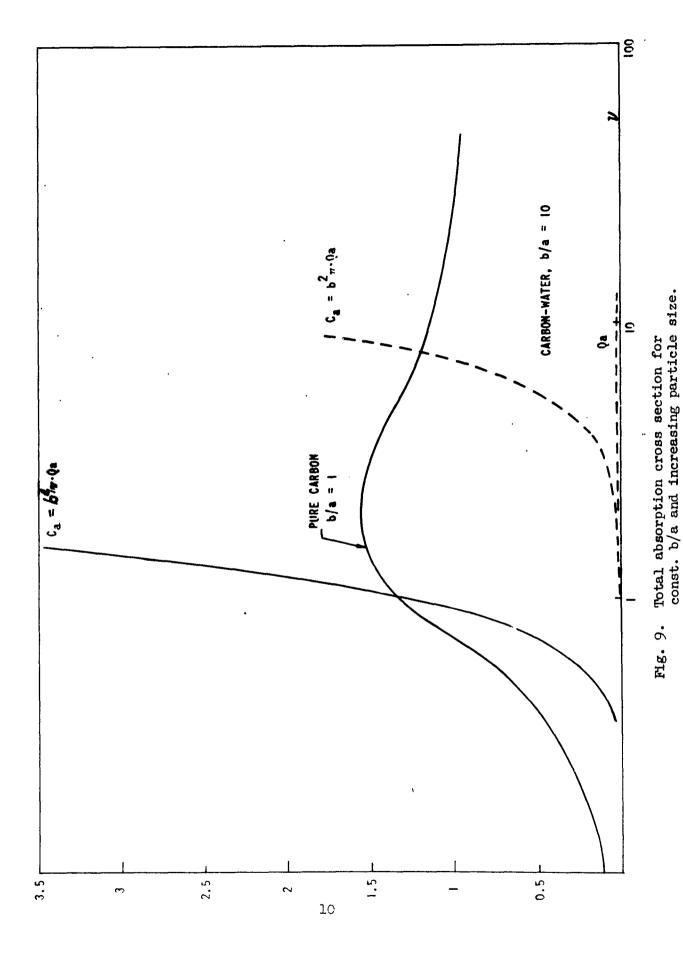
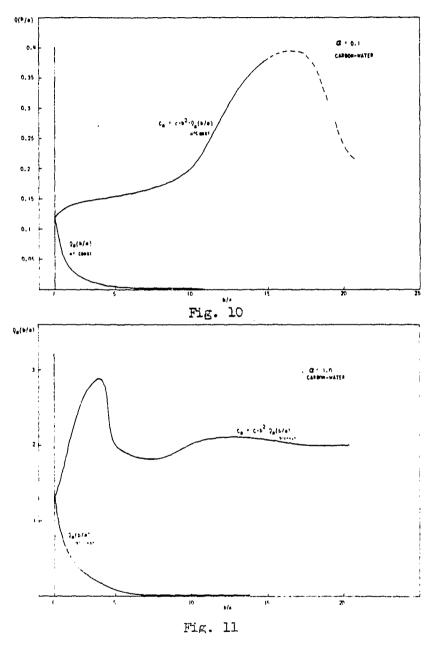


Fig. 8. Absorption coefficient for constant total particle size as function of water shell thickness.





Absorption cross section for constant carbon nucleus size as function of water shell thickness.

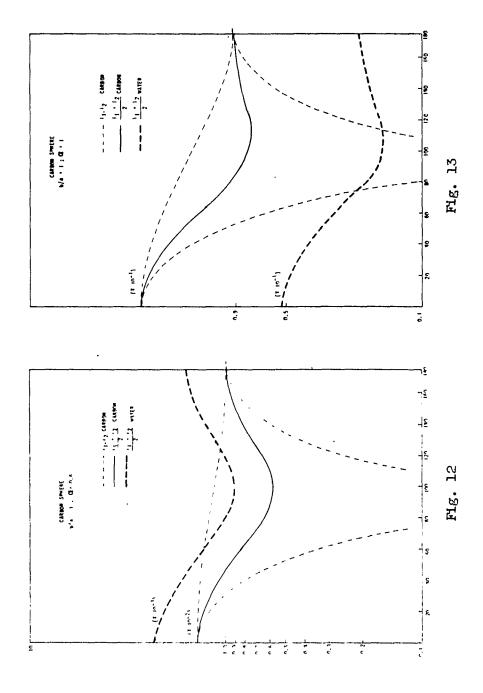
The influence of water absorption in the infrared region on the total absorption of compound carbon-water particles must also be studied. In Table 1 the computed Q_a are listed for a total particle size b=6.265 (to be comparable with Shifrin's computations?) and for carbon-particle radii between a=0.171 and 0.48 for wavelengths from 1.35 to 10 microns. The absorption data for water in these computations have been taken from Dietrich and Shifrin.? It can be seen that the contribution of the carbon particle to the total Q_a decreases with increasing specific ration b/a, and if b/a is larger than about 10, only about 10 percent of the total absorption will be due to the carbon nucleus. In other words, with respect to absorption properties, a compound carbon-water particle with large b/a can be treated in the infrared like a pure water droplet without introducing a large error.

Table 1. Absorption coefficients for water droplets (r = 6.265 µ) with carbon particles of a = 0.171, 0.215, 0.368 and 0.480 µ radius, and for pure water droplets.*

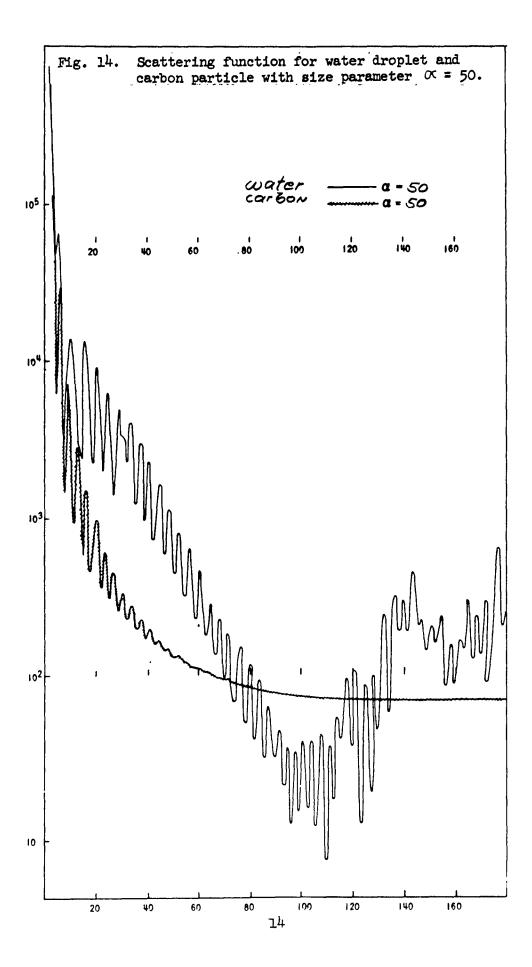
<u> </u>		Qa, water and carbon			Qa, pure water	
	b/a = 16.6 a = 0.171	b/a = 29.1 a = 0.215	b/a = 17 a = 0.368	b/a = 13.5 a = 0.480	b/a = ∞	
1.35 1.5 2.0 3.0 3.2	0.0051 0.0427 0.0790 1.1846 1.1910	0.0062 0.0440 0.0798 1.1846 1.1910	0.0121 0.0493 0.0851 1.1846 1.1910	0.0171 0.0537 0.0899 1.1846 1.1910	0.894	
3.4 4.5 5.47 6.0 7.0	1.0143 0.5004 0.3716 1.1311 0.7374 0.5751	1.0144 0.5005 0.3719 1.1311 0.7376 0.5753	1.0153 0.5022 0.3734 1.1312 0.7384 0.5757	1.0162 0.5045 0.3754 1.1313 0.7395 0.5763	0.925 0.500 1.127 0.704 0.575	

In Figs. 12 through l^4 the scattering functions have been plotted for a pure carbon particle for $\alpha=0.6$ and $\alpha=1$ and also $\alpha=50$. As a comparison, the scattering function for a pure water droplet of the same size has been added. It can be seen that the angular distribution of the scattered light for small α -values depends only slightly on the refractive index. However, the total amount of scattered light is larger for carbon than for water particles. The scattering function for large-size parameter values is particularly interesting since, until recently, no exact computations for large α -values were available. With increasing α , the convergence of the infinite series of the a_n and b_n becomes very poor, and more and more terms have to be computed. In addition, the number of oscillations in the i_1 and i_2 increases proportionally to α . As Van DeHulst 9 has shown, the

^{*}Values for water droplets (r = 6.255 t) taken from Shifrin. 7



Scattering function for carbon particle with size parameter C = 0.6 and |



distance between two consecutive maxima becomes approximately $180^{\circ}/\alpha$. This has been proved by computations of Giese¹⁰ for water drops ($\alpha = 25$, 50, 100). It is also interesting to notice that these oscillations for absorbing particles become completely suppressed for angles $\theta > 50^{\circ}$. The broad maximum between 140° and 145° for water droplets which indicates the fogbow is not present for absorbing particles. It is also important to notice that for such large particles most of the total scattered light is scattered into the narrow forward direction of about zero to 2° , the intensities per unit solid angle being $\approx 10^{\circ}$ times the ones for back-scattering angles. This property is independent of the refractive index and the same for all α above 25. The physical explanation for this is that for large particles the diffraction of light occurs only in these very forward angles. For most measurements it cannot be distinguished from the direct light, and therefore the α for such particles can be assumed as 1 instead of 2.

Absorption of Radiation by Carbon Particles Suspended in the Atmosphere.

Before studying the absorption properties of a carbon-particle cloud, the optimum combination of particle number and size with respect to absorption for carbon particles must be found for a given total carbon mass.

The absorption coefficient of a particle cloud per unit volume is

$$f_{\mathbf{a}}^{\mathbf{r}} = \mathbf{N} \cdot \mathbf{r}^{2} \pi \mathbf{Q}_{\mathbf{a}} (\alpha), \tag{8}$$

where N = number of particles per unit volume. If the volume of one particle is $V_p = {}^{\underline{t}} \mathcal{T} r^3/3$ and the total mass M of particles per unit volume is $V_t \cdot S$ (S = density), the number of particles per unit volume is $F = V_t \cdot 3/4$ $\mathcal{T} r^3 = 3M_t/4$ $\mathcal{T} r^3 \cdot S$. And f_a becomes

$$\mathcal{J}_{\mathbf{a}} = \frac{3}{4} \frac{\mathbf{M}}{9} \frac{1}{r} Q_{\mathbf{a}} (\alpha), \qquad (9)$$

or with $r = \propto \lambda / 2 \pi$,

11

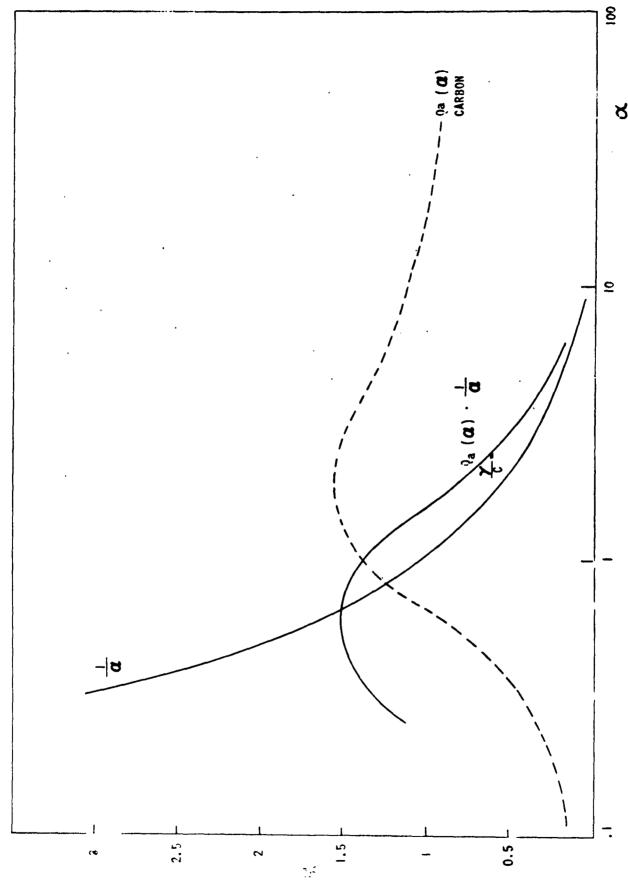
$$f_{\mathbf{a}} = 3\pi \frac{M}{g} \frac{1}{2\lambda \cdot \alpha} Q_{\mathbf{a}} (\alpha)$$

$$f_{\mathbf{a}} = \text{const} \frac{1}{\alpha} Q_{\mathbf{a}} (\alpha). \tag{10}$$

In Fig. 15, the function Q_a (α)/ α is plotted, and the graphical solution gives f_{max}^e for $\alpha = 0.6$.

For sunlight with an intensity peak at $\lambda = 0.5$ micron, $\alpha = 0.6$ corresponds to a = 0.05 micron.

It should be mentioned here that carbon black of this size is commercially available; however, it is very likely that these small particles will form larger conglomerates during the dispersion process. Laboratory experiments on this problem will be necessary.



Total absorption coefficient for a given total carbon mass per unit volume as a function of the size parameter. Fig. 15.

After all necessary properties of single particles were investigated, a study of the processes of such particles in a cloud was begun. The following considerations and computations are based mainly on a thesis by G. Korb (1961, University of Munich, Germany) whose studies were carried out on Signal Corps Contract EUC-1612. The object of this study was to calculate the absorption, transmission, and reflection of radiation propagating through a cloud. In order to understand the significance of the results, a brief summary is given of Korb's solution.

In order to include multiple scattering effects in the cloud, a solution must be found for the general radiation transfer equation (Chandrasekhar¹¹). Korb's approach is to solve the equation for $2 + 2\pi$ radiation fluxes--one upward, one downward and 2π fluxes in the horizontal. This approximation reduces Chandrasekhar's equation to a system of linear differential equations whose solutions give the absorption, transmission, and reflection of radiation. The absorption is given as the amount of energy absorbed inside the cloud by particles and water vapor. The method also takes into consideration the absorption of radiation by water vapor above and below the cloud. These computations of the absorption, transmission, and reflection have to be carried out separately for narrow spectral bands of the sun spectrum (depending on the spectral dependence of the particle parameters), and eventually have to be added up to give the total values. If the albedo of the ground surface is # 0, the same procedure must be applied to so-called auxiliary fluxes, i.e., the diffuse radiation fluxes reflected from the ground surface; and the resulting absorption, transmission, and reflection of the primary and secondary fluxes must be added up.

The first part of Korb's study included the computation of absorption, transmission, and reflection for three model clouds of pure water droplets-a low cloud, a high cloud, and a cloud with large vertical extent.

Korb's method was applied here to calculate the absorption properties of a pure carbon particle cloud, a cloud mixed of droplets and carbon particles, and a cloud of compound concentric carbon-water droplets, all three clouds having a vertical extent of 1,000 meters, with their base near the ground surface:

- (a) Pure carbon particle cloud. The carbon particle concentration is 10^{4} particles per cm³, 0.1-micron-diameter size. This is equivalent to approximately 20 pounds of carbon black per 1 km² for a 1,000-meter-thick cloud. The water-vapor content in the cloud is assumed to be 2 · 10^{-6} g/cm³; the total water-vapor content above the cloud (above 850 mb) is 1.5 cm ppw (according to an average distribution over middle latitudes for summer, Korb, et al¹²).
- (b) Mixed cloud. The cloud consists of droplets of 4 microns radius, $597/\text{cm}^3$, and 10^4 carbon particles/cm³ of 0.05 micron radius. The water-vapor content above the cloud is the same as before--1.5 cm ppw; the saturation water vapor density in the cloud is assumed to be 8 · 10^{-6} g/cm³ at about 47.5° C, and the albedo of the ground is zero.
- (c) Compound particle cloud. This is a cloud of particles which consists of a spherical carbon nucleus with a concentric water shell. The total particle size again is four microns radius and the concentration is

 $597/\text{cm}^3$. (This number results from a water content in the cloud of 0.16 g/m³.) All the carbon particles of the cloud in paragraph (a) will be inside the droplets, which gives 10,000/600 = 16.7 carbon particles/droplet. The size of the combined carbon-particle nucleus becomes approximately 0.125 micron radius, which gives a specific ratio b/a = 32. The water-vapor content in and above the cloud is the same as in paragraph (b).

The sun spectrum from 0.2 through 10 microns has been divided into 17 bands, and for each band the following quantities were computed:

$$k = \frac{K_{a}}{K_{e}} = \frac{\sum n \mathcal{T}r^{2} Q_{a} + S_{w} k_{w} (\lambda)}{\sum n \mathcal{T}r^{2} Q_{e} + S_{w} k_{w} (\lambda)}$$
(11)

and

1

$$T = \int K_e(\lambda) \cdot dh$$
, the optical density of the cloud. (12)

Then the intensity of sun radiation incident on the cloud, E_0 (λ)

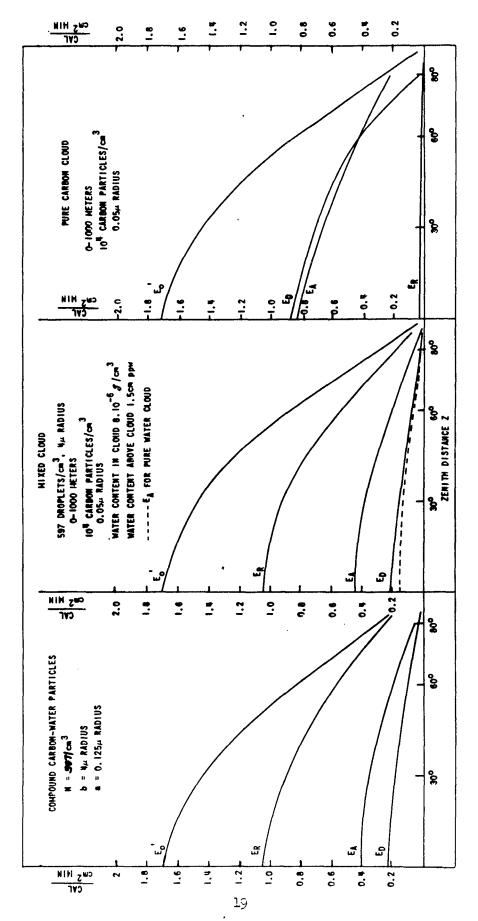
was computed; and finally the absolute values of absorbed, transmitted, and reflected energy were obtained. All computations have been carried out for zenith distances of the sun of 0°, 30°, 60° and 80°.

The results of the computations are plotted in Fig. 16. In the graphs for a mixed cloud, the absorption curve for a pure water-droplet cloud (taken from Korb's report) has been added for comparison. Also added is the curve for E'o, i.e., the intensity of energy incident on the cloud top after passing through the upper atmosphere. Approximately $0.2 \cdot \text{cal/cm}^2 \cdot \text{min.}$ are absorbed by water vapor in the upper atmosphere above the cloud, for vertical incidence. In the case of the pure carbon-particle cloud, the percentage of absorbed and transmitted energy is almost equal, with very little energy (<5 percent) reflected back from the cloud top. For very-low sun elevations, such a cloud absorbs around 80 to 90 percent of the incident radiation, although the absolute amount of absorbed energy becomes very small for low sun elevations. If, as a comparison, the absorption in such a pure carbon cloud for single scattering only is calculated applying Bouguer's law,

$$E_{a} = E_{0} \cdot (1 - e^{-\int a \cdot L}),$$
 (13)

substitution is made for $V_a = N \cdot r^2 T_{Ga} = 5.5 \cdot 10^{-7} \text{ cm}^{-1}$, and the path length for zenith distance z = 0 is 10^5 cm. Then, $E_a/E_0 = 5.5$ percent. The absorption by water vapor in the cloud is approximately 2.5 percent. This would give a total absorption of about 7 percent, only one-sixth of the actual absorption.

The optical properties of the mixed cloud are considerably different. About 60 percent of the incident radiation is reflected from such a cloud so that only a small portion of the radiation penetrates into the cloud. However, it can be seen that, because of the carbon particles, about three times more energy is absorbed than in the pure water-droplet cloud. Only about 12 percent of the incident light is transmitted through this cloud.



Computed transmitted energy (E_{D}) , absorbed energy $(E_{\mathtt{A}})$ and reflected energy $(E_{\mathtt{R}})$ for three cloud models, as function of sun zenith distance. Fig. 16.

A very interesting result is that the optical properties of the mixed and the compound cloud are almost equal, the absorption of the mixed cloud being about 2 to 2.5 percent (of E') higher than for the compound particle cloud. From this it can be concluded that the form in which the carbon particles are distributed in the water-droplet cloud--whether they are in between the droplets, or inside, or on the surface--makes little difference in the absorption coefficient so long as the total carbon mass is about the same. This result is not too surprising if one looks at the spectral distribution of the absorbed energy. About 45 to 50 percent of the total absorbed energy is absorbed in the infrared between two and ten microns wavelength, although there is only about 27 percent of the total incident energy in this wavelength range. This is because of the high absorption of water in the infrared. This wavelength region is therefore influenced very little by the carbon absorption.

In the visible range, Figs. 10 and 11 show that for large b/a the absorption cross section $C_a = b^2 \, \mathcal{T} \cdot Q_a$ remains rather constant and is only slightly higher than the one for the carbon nucleus without water shell. This result is important because it is not actually known what happens to the carbon particles in the cloud after seeding--whether they will attach themselves to the surface of droplets, or whether most of them will remain suspended in the air. It can be assumed that the rather special case of concentric compound particles will exist very seldom--probably only if condensation of water on the carbon particles occurs, most likely by radiational cooling of the carbon particles during nighttime.

So far in these calculations it has been assumed that the albedo of the ground surface is zero. This means that the upward-directed radiation flux at the bottom of the cloud is zero. The more light there is reflected from the ground back into the cloud, the higher the total absorption in the cloud will be. Korb has computed the absorption, transmission, and reflection of pure water-droplet clouds for various albedo values between zero and 0.9, and from his data it can be estimated that in the case of a mixed and compound particle cloud the absorption would be increased by about 2 to 3 percent (of Eo) if the ground albedo were 0.9 instead of zero. The increase in absorption is so small because, in addition to the ground-reflected radiation, a great percentage of radiation is reflected down from the bottom surface of the cloud. This increases the transmissivity of the cloud considerably. For the carbon particle cloud, of course, higher ground albedo would result in a much higher absorption. The denser a cloud, the less influence the ground albedo has on the absorption of the cloud.

For energy processes inside the cloud, the vertical distribution of the total absorbed energy within the cloud must be considered. From Korb's thesis it follows that for a cloud with optical thickness 30 km⁻¹ (as in the compound and mixed-cloud model), for zenith distance $z=30^{\circ}$ and ground albedo $A_{\rm g}=0$ and $K_{\rm a}/K_{\rm e}=0.003$, 50 percent of the total absorbed radiation is absorbed in the upper 300 meters of the cloud; whereby, for 2 < L < 30, the intensity of the downward-directed radiation flux almost follows a distribution of the form E=-a L+b. The denser the cloud, the greater the percentage of absorbed energy in the upper portion of the cloud.

If results of this study are compared with previous calculations by other investigators, the rather detailed study by Smith, et al should be mentioned. They have computed the transmittance, reflectance, and absorption

of a mixed cloud of droplets and small absorbing particles, neglecting the absorption in droplets and applying the two-flux model by S. Fritz. For high optical thicknesses the two-flux model gives about 10 percent lower transmission values, according to comparisons in Korb's report. Using calculations of Smith, Wexler and Glasser, 13 a cloud transmission of about 5 to 10 percent, and a reflectance of about 80 percent, a 10 to 15 percent absorption is obtained for the compound-particle cloud. This is for vertical incidence. Computations made under this study give about 25 percent absorption, 60 percent reflection, and 15 percent transmission. It seems that the two-flux model overemphasizes the reflectance of a cloud. For detailed discussions of the accuracy and significance of data obtained with the 2 + 2 Tflux model, reference is made to Korb's thesis.

Finally, an estimate is made of how much cloud evaporation or dissipation can be achieved with this absorbed energy. The radiation energy which is absorbed within the cloud is used for heating of the total air mass and water-droplet mass, and then for evaporation of the droplets. If it is assumed that the absorption and evaporation process occurs slowly enough so that equilibrium conditions are close between all the components and the atmosphere, then, for the energy budget,

$$dq = c_{pa} \cdot fa \cdot dT + c_{w} \cdot w \cdot f_{w} \cdot dT + L \cdot w.$$
 (14)

cpa = specific heat of air for const. pressure, 0.24 cal/g °C

\$a = density of air, at 7.5° and 90° mb = 1.12 g/cm³

 c_w , f_w = spec. heat and density of water;

w' = liquid water content of cloud g/m^3 ,

L = latent heat of vaporization of water, 590 cal/g °C.

Substituting these values into equation (14),

dq =
$$269 + 0.155 + 87$$
 (cal/m³)
= 356 cal/m³ for a 1°C temperature increase.

Heating of the carbon particles has been neglected completely because their mass is only about 1/100,000th of the water content and it is seen that even the energy required for the heating of the droplets is negligible. The temperature rise which is required to increase the saturation water vapor content from the original 8 g/m^3 to 8.15 g/m^3 is approximately 0.3°C . From equation (14), therefore, is obtained the energy required to evaporate the compound-particle cloud and to achieve the necessary temperature increase of 0.3°C to about 100 cal/m^3 .

In the case of the mixed cloud and the compound-particle cloud, the absorbed energy for sun zenith distances between zero and 30° is about 0.4 cal/cm² min. The air column below l cm² is l \cdot 105 cm³ = 0.1 m³, which gives an average absorbed energy of 4 cal/m³ \cdot min. This would result in a necessary absorption time of about 25 minutes. For a sun zenith distance of 60°, the absorbed energy would be only about 0.2 cal/cm² min. and the absorption time therefore twice as long--approximately 50 minutes.

Actually, it must be expected that the upper 100 meters of the cloud, where the absorption per unit volume is about twice the average absorption, will start to warm up and evaporate first. This will, again, allow more radiation to penetrate into the lower layers of the cloud. However, as soon as the upper layer starts to warm up it will also begin to rise and cool off again. It certainly will be necessary to provide more than the minimum energy for a 0.3°C temperature increase if the evaporation process is to be maintained long enough to evaporate the cloud completely.

Van Straten⁵ suggested as a hypothetical process for the cloud dissipation an evaporation and condensation process between droplets. Droplets containing many carbon particles would heat up more than pure water droplets, and therefore moisture would evaporate from the warmer droplets with the carbon particles onto the colder pure water droplets.

A droplet containing carbon particles will absorb energy and consequently its temperature will rise. This again will cause an energy loss by conduction and evaporation. The energy budget can be written in the following way:

$$\frac{dE_a}{dt} = c \cdot m \cdot \frac{dT}{dt} + 4 \mathcal{T} r \cdot K \cdot (T - T_0) + 4 \mathcal{T} r^2 \cdot S \cdot L \frac{dr}{dt}$$
(15)

c = specific heat of water = 1 cal. degr. $^{-1} \cdot g^{-1}$

m = mass of droplet

r = droplet radius

13

 $K = \text{thermal conductivity of air} = 5 \cdot 8 \cdot 10^{-5} \text{ cal. cm}^{-1} \cdot \text{sec}^{-1} \cdot \text{degr}^{-1}$

T = droplet temperature

 T_0 = air temperature $S' = \text{water density} = 1 \text{ g} \cdot \text{cm}^{-3}$ $L = \text{latent heat of evaporization} = 590 \text{ cal} \cdot \text{g}^{-1} \cdot \text{degr}^{-1}$

whereby $\frac{dr}{dt} = \frac{D}{\epsilon \cdot r} (s_d - s_w)$ (16)

D = coefficient of diffusion of water vapor = $0.226 \text{ cm}^2 \cdot \text{sec}^{-1}$ S_{d} , S_{W} = vapor density of water at the droplet surface and far away from the droplet, respectively.

Id is a function of T and r, according to Kelvin's formula:

$$\mathcal{S}_{d} = \frac{M}{R \cdot T} \cdot P_{w}(T) \cdot \exp(\frac{2\sigma M}{R \cdot r \cdot T})$$
 (17)

 σ = specific surface energy of water = 74.5 cal \cdot cm⁻²

M = molecular weight of water = 18

R = universal gas constant = 1.98 cal \cdot g⁻¹ \cdot degr⁻¹.

Substituting (16) and (17) in (15),

$$\frac{dE_{p}}{dt} = c \cdot m \cdot \frac{dT}{dt} + 4 \pi \cdot K(T-T_{0}) + 4 \pi LD \left[\left(S_{w}(T) \right) \right] \cdot \exp\left(\frac{2\sigma M}{RrT} \right) = S_{w}(T_{0}). \quad (18)$$

As a first approximation, it is assumed that r is constant and the following abbreviations are used:

with $Q_a = 0.002$, $E_0 = 1.5 \text{ cal} \cdot \text{cm}^{-2} \cdot \text{min}^{-1}$.

We then get

We then get
$$q = \alpha \frac{dT}{dt} + \beta (T-T_0) + \beta^2 \left\{ S_w (T) e^{\frac{S}{T}} - S_w (T_0) \right\},$$
or
$$\alpha \frac{dT}{dt} + \beta T + \beta S_w (T) e^{\frac{S}{T}} = q + \beta T_0 + \beta S_w (T_0) \qquad (20)$$

c = q + $\mathcal{G}T_0$ + $\mathcal{G}W_0$ (T₀) is constant. The magnitude of each of the three quantities (q = 2.5 · 10⁻¹¹, $\mathcal{G}T_0$ = 8 · 3 · 10⁻⁵, and \mathcal{G} · $\mathcal{S}_W(T_0)$ = 5.4 · 10⁻⁶), shows that q will have to be 100,000 times bigger to be of any influence on the result of this equation. This confirms Smith, Wexler, and Glaser's conclusion that 104 particles per droplet would be necessary to achieve a sufficient rise of temperature in the droplet.

An analytical integration of equation (18) will be extremely difficult, if not impossible. However, since in this study there is not much interest in the transient phase, dT/dt = 0 is assumed and T is calculated only after

reaching equilibrium conditions.

$$(\mathfrak{T} + \mathfrak{f}^{\mathfrak{S}}_{W} (\mathfrak{T}) e^{\overline{\mathfrak{T}}} - c = 0. \tag{21}$$

The numerical solution gives T = 280.46°K or 7.3°C. The temperature of the particle has decreased by 0.2°C from the initial temperature, To = Tair. From an assumed air temperature of 7.5°C, evaporation 'ue to the curvature of the droplet was obtained. Also, from equation (18), it can be seen that the condensation term is of the order 10-7 and the evaporation term is of the order 10⁻⁰; and the first term, the internal energy increase, is of an order $\angle 10^{-10}$, which means that most of the absorbed energy is lost by conduction to the surrounding air. This loss of energy to the air will actually be even greater since the heat transfer does not occur only by conduction but also by convection. Consequently, the air temperature will rise almost as fast as the particle temperature. It has been shown that for small absorption rates the particle temperature may be even below the air temperature because of evaporation due to vapor-pressure difference. Most of the

evaporated water from the droplets will be needed to maintain saturation in the heated air, and only a very small portion would be available for condensation on pure droplets. Therefore, it can be stated definitely that an evaporation-condensation process cannot be considered as a possible cloud-dissipation process, and therefore the only possible dissipation process for carbon-seeded clouds will be evaporation due to heating of the total air mass which, as has been shown, requires very large quantities of carbon black.

This report has been limited to absorption processes only, and no discussion has been made of any emission effects from carbon seeded clouds. For more information on the energy budget in clouds, a study is certainly necessary. As a result of the very encouraging results of the first part of Korb's study and the results reported here, the studies by the University of Munich, Germany, have been extended under Signal Corps contract to also cover various models of pure and mixed carbon and water clouds to a much hashed extent and accuracy than has been done in work reported here. This extended study will also include energy losses due to emission from the cloud.

CONCLUSIONS

1

A summary of the results indicates that for a model cloud of approximately 600 droplets/cm³, 4 micron radius, from ground to 1,000 meters altitude (stratus-type cloud), seeding with 20 pounds of 0.05 micron radius carbon particles will increase the absorption of sun radiation within the cloud from about 0.15 to 0.4 cal/cm² \cdot min for 0° to 30° sun zenith distance.

The absorption, transmission and reflection are almost equal for a mixed cloud of water droplets and carbon particles and a cloud of compound concentric carbon-water particles. This indicates very strongly that for absorption studies it is not very critical to know in what form the carbon particles are distributed within the cloud, i.e., whether they are contained within the cloud droplet or attached to its surface.

From estimates of the energy required for evaporation of the model cloud it is concluded that for realistic field experiments it will be necessary to use seeding rates of the order of at least 100 pounds of carbon black per km² during high sun elevations and for relatively stable cloud formations. The necessary seeding rate may go up to several hundred pounds per km² under less favorable conditions.

An analysis of the energy budget in the seeded cloud yields that an evaporation-condensation process will not be possible, and that evaporation of a cloud due to heating of the total air mass will be possible only under very favorable conditions.

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The temperature change of one particle caused by radiation absorption has been computed, and the total energy for evaporation of a cloud mass has been estimated.

From these calculations it can be concluded that a total carbon mass of the order of several hundred pounds per km^2 will be necessary to evaporate a stratus-type cloud.

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